

Phonon-assisted carrier motion on the Wannier-Stark ladder

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It is well known that at zero temperature and in the absence of electron-phonon coupling, the presence of an electric field leads to localization of carriers residing in a single band of finite bandwidth. We implement the Self-Consistent Born Approximation (SCBA) to study the effect of weak electron-phonon coupling on the motion of a carrier in a biased system. At moderate and strong electron-phonon coupling we supplement the SCBA, describing the string of phonons left behind by the carrier, with the Momentum Average (MA) approximation to describe the phonon cloud that accompanies the resulting polaron. We find that coupling to the lattice delocalizes the carrier, as expected, although long-lived resonances resulting from the Wannier-Stark states of the polaron may appear in the spectrum in certain regions of the parameter space. The approach we propose here can also be used to implement and check the validity of simple variational approximations.

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I. INTRODUCTION

It has long been known^{1,2} that carriers in a clean one-dimensional tight-binding band become localized³ if a uniform electric field E is applied, since this breaks the free-particle continuum into a sequence of equally spaced discrete levels separated by the electric potential energy between consecutive sites $\delta = eaE$, where e is the carrier's charge and a the lattice constant. This discrete spectrum is the Wannier-Stark (WS) ladder.⁴

While the ladder has been observed in semiconductor superlattices and in cold-atom systems,² it is not seen in the spectra of regular crystalline solids. The absence of localization is easily understood in metals, because the Fermi sea electrons screen out the electric field and carriers move ballistically as described by the Buttiker-Landauer theory⁵ (if correlations can be neglected). In insulators, however, the electric field is not screened out and therefore the band is "tilted". Here, the absence of localization is attributed to coupling to the lattice: a carrier can emit phonons⁶ and thus lower its energy to slide along the chain, as sketched in Fig. 1. Most previous work on this problem assumes incoherent tunneling between sites.⁶ For example, this is routinely done when modeling carrier transport in organic solar cells, based on the belief that those organic semiconductors are so disordered as to destroy coherence.⁷ While this assumption awaits validation, an understanding of the full quantum dynamics, which should be relevant in clean(er) systems,⁸ is still needed.

The quantum problem was first studied numerically in Ref. 9, with a variational solution assuming that phonons appear only on the same site or to the left (uphill) of the carrier. Ref. 10 obtained analytic and numerical results for the spectrum of a finite chain for weak electron-phonon (e-ph) coupling and small hopping $t \ll \delta$, while Ref. 11 investigated the time evolution of the wavefunction once the electric field is turned on.

The method we propose here is similar in spirit to that used in Ref. 9, however we use a different assumption to calculate analytically the Green's function for this problem. Unlike Ref. 9, we do not restrict the direction of motion of the carrier, instead we assume that the phonons left behind by the carrier can only be absorbed in inverse order to that in which they were emitted. This leads to only non-crossed diagrams being summed in such processes, which is the essence of the Self-Consistent Born Approximation (SCBA). For the non-biased system ($\delta = 0$), SCBA is known to be accurate only at weak e-ph coupling. For moderate and strong e-ph coupling, we use SCBA to describe this string of phonons left behind as

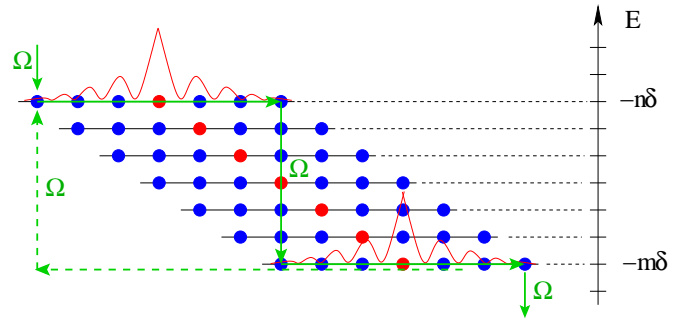


FIG. 1. (color online) Carrier motion on the WS ladder. The horizontal axis is the chain, with sites shown as dots. The vertical axis is the energy. Several WS eigenstates are shown, each centered at the site (red dot) with the same on-site energy. The probability distribution is sketched for two of these. Green arrows show part of the evolution, with the carrier arriving on the $-n\delta$ level from a higher one upon phonon emission; it then hops towards right and eventually emits another phonon to move on the $-m\delta$ level, etc. If phonons are absorbed in reverse emission order when the carrier retraces its steps, then only non-crossed diagrams are generated. Crossed diagrams are for processes shown by the dashed line, where phonons are not absorbed in reverse emission order.

the carrier moves to lower energies, and combine it with the Momentum Average (MA) approximation to describe the phonon cloud that accompanies the carrier, turning it into a polaron. MA has been shown to provide a rather accurate description of the polaron properties for any e-ph coupling strength in an unbiased system, so long as the energy of the optical phonons, Ω , is not too small.¹²

We argue that taken together, these approximations allow us to understand the local density of states (LDOS) in such a system if the bias δ is not large compared to Ω . Our results uncover the evolution of the LDOS as the e-ph coupling is turned on, confirming that delocalization occurs as soon as such coupling is present. However, for strong e-ph coupling and smaller biases, very sharp resonances can appear in the spectrum, and are understood as being due to WS-like states for the polaron, which however can tunnel into extended states located further downhill. We believe that these results supplement those presented in Refs. 9–11 to improve our understanding of the quantum dynamics in this system. The formalism we propose here can also be easily modified to implement other variational descriptions to check for their validity, as we exemplify for two particular cases. Other possible generalizations are discussed at the end.

The article is organized as follows: Section II describes the model and the formalism we use to calculate the propagators and resulting LDOS. The results are presented in Section III, while Section IV contains a summary and some further discussions.

II. MODEL AND FORMALISM

The model Hamiltonian we study is described by:

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_{ph} + V_{e-ph} \quad (1)$$

where

$$\mathcal{H}_e = -t \sum_n (c_n^\dagger c_{n+1} + h.c.) + \sum_n \epsilon_n c_n^\dagger c_n$$

describes nearest-neighbor hopping of the carrier on a 1D chain biased by the applied electric field, so that the on-site energies are $\epsilon_n = -neaE = -n\delta$. (The spin is trivial and we ignore it for simplicity). There is an Einstein phonon mode

$$\mathcal{H}_{ph} = \Omega \sum_n b_n^\dagger b_n$$

(for simplicity, we take $\hbar = 1$ in the following). Finally,

$$V_{e-ph} = g \sum_n c_n^\dagger c_n (b_n^\dagger + b_n)$$

is the Holstein model¹³ for e-ph coupling. As usual, c_n and b_n are annihilation operators for the carrier and phonons, respectively, at site n of the chain. Also as customary, we will gauge the strength of the e-ph coupling with the dimensionless effective coupling:

$$\lambda = \frac{g^2}{2t\Omega}$$

appropriate for 1D models.¹²

The quantity of interest is $G(n, z) = \langle 0 | c_0 \hat{G}(z) c_n^\dagger | 0 \rangle$ where $|0\rangle$ is the vacuum and $\hat{G}(z) = [z - \mathcal{H}]^{-1}$ is the resolvent at $z = \omega + i\eta$, where $\eta \rightarrow 0$ controls the artificial lifetime $\sim 1/\eta$ of the carrier. This is the Fourier transform of $G(n, \tau) \sim \Theta(\tau) \langle 0 | c_0 e^{-i\mathcal{H}\tau} c_n^\dagger | 0 \rangle$, *i.e.* the amplitude of probability for the carrier to move from site n to site 0 in a time τ and so that all phonons emitted in the meantime have been re-absorbed. If such a process is very unlikely, then $G(n, z) \rightarrow 0$. From the Lehmann representation¹⁴ we know that the local density of states (LDOS) $A(n, \omega) = -\frac{1}{\pi} \text{Im} G(n, z)$ is finite at energies $\omega = E_\alpha$ in the one-carrier spectrum $\mathcal{H}|\phi_\alpha\rangle = E_\alpha|\phi_\alpha\rangle$, provided that the overlaps $\langle 0 | c_0 |\phi_\alpha\rangle \langle \phi_\alpha | c_n^\dagger | 0 \rangle$ do not vanish. As will become apparent soon, our method to calculate $G(n, z)$ also gives the generalized propagators:

$$F_k(n; n_k, \dots, n_1; z) = \langle 0 | c_0 \hat{G}(z) c_n^\dagger b_{n_k}^\dagger \dots b_{n_1}^\dagger | 0 \rangle \quad (2)$$

whose meaning and usefulness mirror those of $G(n, z)$.

A. No e-ph coupling: $\lambda = 0$

We first calculate $G_0(n, z) = \langle 0 | c_0 \hat{G}_e(z) c_n^\dagger | 0 \rangle$ for \mathcal{H}_e , *i.e.* when there is no e-ph coupling. Taking appropriate matrix elements of the identity $\hat{G}_e(z)(z - \mathcal{H}_e) = 1$ gives the equations of motion (EOM):

$$(z - \epsilon_n) G_0(n, z) = \delta_{n,0} - t[G_0(n-1, z) + G_0(n+1, z)] \quad (3)$$

These are solved easily if we recognize that $G_0(n, z) \rightarrow 0$ for sufficiently large $|n|$ because the electron cannot move arbitrarily far in a finite lifetime $1/\eta$. As a result,

$$\begin{aligned} G_0(n, z) &= A(z - \epsilon_n) G_0(n-1, z) \quad , \text{ if } n > 0 \\ G_0(n, z) &= B(z - \epsilon_n) G_0(n+1, z) \quad , \text{ if } n < 0 \end{aligned} \quad (4)$$

where we define the continued fractions:

$$\begin{aligned} A(f(z)) &= \frac{-t}{f(z) + tA(f(z + \delta))} \\ B(f(z)) &= \frac{-t}{f(z) + tB(f(z - \delta))}. \end{aligned} \quad (5)$$

These quantities are calculated iterationally starting from a cutoff $A(z + N\delta) = B(z - N\delta) = 0$ for a sufficiently large N . Because for $\delta \neq 0$ all eigenstates are localized, a cutoff $N \sim 20$ usually suffices. If the electric field is turned off, $\delta = 0$, they can be found analytically to be $A(z)|_{\delta=0} = -B(-z)|_{\delta=0} = -z/2t + \sqrt{z/2t + 1}\sqrt{z/2t - 1}$ so that $|A(z)| < 1$, $|B(z)| < 1$, if $\text{Im}(z) = \eta > 0$. Finally, using the $n = \pm 1$ results of Eq. (4) in Eq. (3) leads to:

$$\begin{aligned} G_0(0, z) &= \frac{1}{z + t[A(z + \delta) + B(z - \delta)]} \quad , n = 0 \\ G_0(n, z) &= A(z + n\delta) \dots A(z + \delta) G_0(0, z) \quad , n > 0 \\ G_0(n, z) &= B(z - n\delta) \dots B(z - \delta) G_0(0, z) \quad , n < 0. \end{aligned}$$

If $\delta = 0$ this gives the usual results for a tight-binding model.¹⁵ For $\delta \neq 0$ it is easy to check that the WS energies $E_n = n\delta$ are indeed poles of $G_0(n, z)$. Full mapping onto the analytic solution¹⁶ can also be verified.¹⁷

B. Weak e-ph coupling, $\lambda \ll 1$: SCBA

For $g \neq 0$, the EOM acquire additional terms because of phonon emission and absorption. In particular, now:

$$(z - \epsilon_n)G(n, z) = \delta_{n,0} - t[G(n-1, z) + G(n+1, z)] + gF_1(n; n; \omega) \quad (6)$$

Exact EOM for $F_k, k \geq 1$ of Eq. (2) can be easily derived, however the resulting infinite system of coupled equations is too complicated, thus approximations are needed.

Physically, we expect the carrier to leave phonons behind, as sketched in Fig. 1, in order to move down the ladder. The more probable processes, leading to diagrams with the largest contributions, are like those shown by full lines: phonons are emitted when needed to move between different ladder states and are absorbed in reverse order if the carrier goes back. A process leading to a crossed diagram is shown by the dashed lines, and should have a low probability because the ladder states are localized. Note that here we assume that the phonons left behind are typically not spatially very close to one another. This is a reasonable assumption if $\Omega > \delta$. Below, we will also gauge the validity of this assumption in the case where $\Omega \sim \delta$.

The assumption that the contribution of crossed diagrams can be ignored is the essence of the Self-Consistent Born approximation (SCBA). For weak coupling $\lambda \ll 1$, SCBA is known to be a reasonable approximation in the un-biased system with $\delta = 0$.¹² This is another reason to expect that its generalization to the biased case, provided here, should continue to work well for small λ .

By keeping only non-crossed diagrams, SCBA assumes that phonons are absorbed in inverse order to their emission order, *i.e.* if phonons were priorly emitted (in this order) at sites n_1, \dots, n_k , at this point either another phonon is emitted, or only the one at n_k can be absorbed. For this to be possible, these phonons must be distinguishable. This is automatically the case if they are located at different sites. If there are multiple phonons emitted at the same site, we will treat them as if they belong to different phonon modes so that they continue to be distinguishable. As we show below, this is implicitly assumed to be true for SCBA in the unbiased system with $\delta = 0$. It should remain a reasonable assumption for the biased case as well if $\Omega > \delta$, since, as already discussed, we do not expect multiple phonons to be located at the same site with high probability. Below we provide a way to gauge the validity of this approximation.

After imposing these restrictions, the EOM for the generalized propagator $F_k(n; n_k, \dots, n_1; z)$, $k \geq 1$, read:

$$(z - \epsilon_{n_k} - k\Omega)F_k(n_k; n_k, \dots, n_1; z) = -t[F_k(n_k - 1; n_k, \dots, n_1; z) + F_k(n_k + 1; n_k, \dots, n_1; z)] + gF_{k-1}(n_k; n_{k-1}, \dots, n_1; z) + gF_{k+1}(n_k; n_k, n_k, n_{k-1}, \dots, n_1; z) \quad (7)$$

if $n = n_k$, while for $n \neq n_k$:

$$(z - \epsilon_n - k\Omega)F_k(n; n_k, \dots, n_1; z) = -t[F_k(n-1; n_k, \dots, n_1; z) + F_k(n+1; n_k, \dots, n_1; z)] + gF_{k+1}(n; n, n_k, \dots, n_1; z). \quad (8)$$

In other words, if the carrier is at the site $n = n_k$ where the last emitted phonon resides, it can hop away, it can absorb that phonon or it can create another phonon (treated as if it belongs to a different mode) at the same site. If the carrier is at a site $n \neq n_k$ it can hop away or emit a new phonon, but absorption of one of the existing phonons is not allowed by the non-crossing condition.

Remarkably, these EOM can be solved analytically by noting that for any $k \geq 0$ we must have

$$F_{k+1}(n; n, n_k, \dots, n_1; z) = \sigma(z - \epsilon_n - k\Omega)F_k(n; n_k, \dots, n_1; z). \quad (9)$$

Mathematically, this is because if k and n are large enough, these propagators must eventually vanish. Truncating the EOM at any $k+2$ (k can be arbitrarily large) leads to a form similar to Eq. (9). This ansatz turns Eq. (8) into a simple recurrence equation like Eq. (3), thus $F_k(n_k + 1; n_k, \dots, n_1; z) = A(z - \epsilon_{n_k+1} - k\Omega - g\sigma(z - \epsilon_{n_k+1} - k\Omega))F_k(n_k, n_k, \dots, n_1; z)$, and $F_k(n_k - 1; n_k, \dots, n_1; z) = B(z - \epsilon_{n_k-1} - k\Omega - g\sigma(z - \epsilon_{n_k-1} - k\Omega))F_k(n_k, n_k, \dots, n_1; z)$. Using these in Eq. (7) leads to an equation consistent with the ansatz of Eq. (9), from which we find:

$$\sigma(z) = \frac{g}{z - \Omega - g\sigma(z - \Omega) + tA(z + \delta - \Omega - g\sigma(z + \delta - \Omega)) + tB(z - \delta - \Omega - g\sigma(z - \delta - \Omega))}. \quad (10)$$

The solution of this equation can be calculated iteratively starting from $\sigma(z) \approx 1/(z - \Omega)$ as $|z| \rightarrow \infty$.

Physically, Eq. (9) means that the amplitude of probability for an additional phonon to be emitted depends only on the energy of the electron, and not on the detailed

locations of the previously emitted phonons. Using the ansatz for $k = 1$ into Eq. (6) leads to:

$$G(n, \omega) = G_0(n, \omega - g\sigma(\omega)) \quad (11)$$

and we recognize $\Sigma_{\text{SCBA}}(\omega) = g\sigma(\omega)$. It is straightfor-

ward to verify that for $\delta = 0$, this is the expected solution $\Sigma_{\text{SCBA}}(\omega) = \frac{g^2}{N} \sum_q G_{\text{SCBA}}(k - q, \omega - \Omega)$ where $G_{\text{SCBA}}(k, \omega) = 1/[\omega + i\eta - \epsilon_k - \Sigma_{\text{SCBA}}(\omega)]$.¹² One can now obtain the SCBA values for other propagators F_k .

To check the validity of this approximation, we can use the same framework to implement other variational schemes and compare the results. For example, the solution of Ref. 9 can be trivially implemented by setting in the EOM $F_k(n; n_k, \dots, n_1, \omega) = 0$ if $n < n_k$, i.e. the carrier cannot be to the left of the last emitted phonon (this automatically implies $n_1 \leq \dots \leq n_k$). As a result, the corresponding self-energy (which we label as “Ref. [9]” in the following) has $B \equiv 0$ in Eq. (10). A priori, we do not expect this approximation to be that good for very small biases where the effective probabilities for the carrier to hop uphill vs. downhill are not that different.

A wider variational space can be achieved by allowing the electron to go anywhere but keeping the additional restriction $n_1 \leq \dots \leq n_k$, i.e. the electron can move to the left of existing phonons but it cannot emit additional phonons while there. Since this is one of the ways to obtain multiple phonons at the same site, this approximation allows us to gauge the importance of contributions from configurations with multiple phonons at the same site. Mathematically, the corresponding EOM for this variational approximation (which we label “var” in the following) are obtained by removing the last term in Eq. (8) when $n < n_k$. Its solution is like Eq. (10) but with $B(z - \delta - \Omega)$ in the denominator (since no new phonons are emitted when the carrier moves to the left of existing phonons, the contribution from such paths is not renormalized by the self-energy $g\sigma$). Various other possibilities can be implemented similarly, by only keeping terms in the EOM consistent with those assumptions, but we stop here.

C. Moderate and large e-ph coupling: MA+SCBA

For stronger electron-phonon coupling, the probability to have multiple phonons at the same site must increase. This is known to be the case even for the unbiased system, because the electron creates a robust phonon cloud that accompanies it as it moves through the system. The resulting dressed quasiparticle is, of course, the polaron. In the biased system, one would expect the polaron to move down the ladder with its robust cloud.

In such conditions, we expect that the approximation made above, of treating multiple phonons that happen to be at the same site as if they belong to different modes, to become quantitatively inaccurate because of normalization factors. To see why, consider a state with n bosons at the same site. If they belong to the same

mode, it is described by $|n\rangle = b^{\dagger,n}/\sqrt{n!}|0\rangle$, and we have $b|n\rangle = \sqrt{n}|n-1\rangle$, etc. However, if we treat the n bosons as belonging to n distinct modes with one boson each, then there are no normalization factors. For small $n \sim 1$ this makes little difference, but this is no longer the case if many bosons are likely to occur at the same site.

Thus, at moderate and large λ , the e-ph coupling has two consequences: one is to lead to the formation of the polaron with its robust cloud, and the other is to allow it to move to lower energies by leaving phonons behind. The number and typical locations of the phonons left behind is controlled by the ratio Ω/δ and therefore is not very sensitive to the strength of the coupling. As a result, we expect these processes to continue to be well described by the SCBA scheme, i.e. by assuming that these phonons are only involved in non-crossed diagrams.

However, at any point the electron can start building a larger cloud in its vicinity (the polaron cloud). Since this cloud typically contains many phonons, it is unlikely that the electron will abandon it and move away to start building another robust cloud, at least not in the case $\Omega \gtrsim \delta$ that we consider here. Instead, the electron will reabsorb these cloud phonons and then move to another location (maybe leaving one phonon behind) and start creating another robust polaron cloud, similarly to how it moves in an unbiased system.

In the unbiased system and for moderate and large e-ph coupling, it has been shown that the Momentum Average (MA) approximation¹² provides an accurate description of the polaron properties so long as Ω is not very small. For the Holstein model, MA has been shown to correspond to the variational approximation of assuming that the polaron cloud has all its phonons at one site.¹⁸ This variational space can be enlarged systematically to check its validity. While for the Holstein model this approximation is already very reasonable, for more complicated models of e-ph coupling one needs to allow the polaron cloud to spread over multiple adjacent sites.¹⁹

Here we implement an MA+SCBA approximation which assumes that a one-site polaron cloud can only be built at the location of the last emitted phonon, and that while a cloud with two or more phonons is present the electron will not emit/absorb phonons in other locations, consistent with MA. At the same time, phonons not in the cloud can only be absorbed in the inverse order to that in which they were emitted, as described by SCBA.

Mathematically, we implement this as follows. The EOM remain unchanged as long as $n_k \neq n_{k-1}$, i.e. no cloud is being built. Equations (8) and (9) are supplemented with additional equations for the propagators with multiple phonons at site n_k . Specifically, for any $p \geq 1$, and using the shorthand notation $\{n\}_{p+1} \equiv n_k, \dots, n_k, n_{k-1}, \dots, n_1$ where the first $p+1$ sites are all n_k , we have:²⁰

$$(z - \epsilon_{n_k} - (k+p)\Omega)F_{k+p}(n_k; \{n\}_{p+1}; z) = -t[F_{k+p}(n_k - 1; \{n\}_{p+1}; z) + F_{k+p}(n_k + 1; \{n\}_{p+1}; z)] \\ + (p+1)gF_{k+p-1}(n_k; \{n\}_p; z) + gF_{k+p+1}(n_k; \{n\}_{p+2}; z) \quad (12)$$

for $n = n_k$, while for $n \neq n_k$:

$$(z - \epsilon_n - (k + p)\Omega)F_{k+p}(n; \{n\}_{p+1}; z) = -t[F_{k+p}(n-1; \{n\}_{p+1}; z) + F_{k+p}(n+1; \{n\}_{p+1}; z)] \quad (13)$$

These additional equations can be solved trivially and give:

$$F_{k+1}(n_k; n_k, n_k, n_{k-1}, \dots, n_1; z) = \sigma_{MA}(z - \epsilon_{n_k} - (k+1)\Omega)F_k(n_k; n_k, \dots, n_1; z) \quad (14)$$

where

$$\sigma_{MA}(z) = \frac{2g}{z + tA(z + \delta) + tB(z - \delta) - \frac{3g^2}{z - \Omega + tA(z - \Omega + \delta) + tB(z - \Omega - \delta) - \frac{4g^2}{\dots}}} \quad (15)$$

The ansatz of Eq. (9) remains unchanged if the last two phonons are not at the same site, and is supplemented by Eq. (14) if the last two phonons are at the same site. The rest of the solution proceeds as before and we find

$$\sigma(z) = \frac{g}{z - \Omega - g\sigma_{MA}(z - \Omega) + tA(z + \delta - \Omega - g\sigma(z + \delta - \Omega)) + tB(z - \delta - \Omega - g\sigma(z - \delta - \Omega))}. \quad (16)$$

Again, we will check this approximation against the variational predictions that do not allow the electron to move to the left of the rightmost phonon (labeled as “Ref. [9]”), respectively allow it to do so but not to emit additional phonons to the left of the rightmost one (labeled as “var”). These are implemented just as before. Another approximation we implement, which will be labeled as “MA+SCBA dressed”, is obtained by replacing $A(z) \rightarrow A(z - g\sigma(z))$, $B(z) \rightarrow B(z - g\sigma(z))$ everywhere in Eq. (15). As its name suggests, this approximation allows the electron to start building additional non-crossed strings of phonons, which may include one-site larger clouds, while the original cloud is present, because it is obtained by replacing bare propagators by full propagators in Eq. (15). Comparing it to the MA+SCBA results will allow us to gauge whether the assumption that such processes can be ignored is correct.

III. RESULTS

A. Weak coupling limit: SCBA

We begin by analyzing a system with a small bias and small e-ph coupling, using the SCBA approximation. Typical results are shown in Fig. 2, which plots the $n = 0$ LDOS for various values of g , with (thin black line) and without (thick red line) an electric field $\delta = 0.1$ for $\Omega = t = 1$. The LDOS at other sites is given by $A(n, \omega) = A(0, \omega - \epsilon_n)$, i.e. it is shifted by $n\delta$.

Figure 2(a) shows the $g = 0$ results. As expected, the biased system’s LDOS shows discrete peaks at $\omega = m\delta$ marking the WS ladder. Some of these peaks are hard to see because their wave-function is very small at site $n = 0$. This is progressively the case for peaks with energies $|\omega| > 2.5t$ because of their localized nature. The LDOS of the unbiased system is the usual 1D result, with a continuum of states for $|\omega| \leq 2t$.

As we turn the e-ph coupling on in panels (b)-(d), the former WS states acquire a finite lifetime (their width is no longer controlled by η , instead being significantly larger even for $g = 0.1$, see change in the vertical scale), showing that these states are no longer localized. This proves that coupling to the lattice indeed results in delocalization. As λ increases, the peaks continue to broaden and start to merge into a smooth continuum. This occurs in an asymmetric way, with higher energy states converging faster towards a smooth LDOS, while the lower energy states still show considerable LDOS variation.

This may seem surprising at first, but the reason becomes clear when we compare with the LDOS for $\delta = 0$ (thick red line), which has two features: a polaron band at low energies $\omega \in [E_{GS}, E_{GS} + \Omega]$ (E_{GS} is the polaron ground-state energy) and the polaron+one-phonon continuum for $\omega > E_{GS} + \Omega$.¹⁸ Arrows mark the transition between the two features, which is barely visible on this scale for $g = 0.1, 0.2$ (for the later case, it is shown more clearly in the inset). States in the polaron band describe the coherent, infinite-lifetime quasiparticle (the polaron) consisting of the carrier and its phonon cloud. In contrast, the polaron+one-phonon continuum contains incoherent states with finite lifetime, describing the scattering of the polaron on one or more phonons that do not belong to its cloud.

At first, one may expect that turning on an electric field should have a very different effect on the two types of states: the incoherent states at high energy should remain delocalized since the polaron already has enough energy to leave phonons behind and can continue to do so when the bias is applied. However, at low energies one may expect to see a WS ladder describing the localization of the polaron. Indeed, if here the polaron carries all the phonons in its cloud then it cannot leave any of them behind, therefore the electric field should localize it just like it does with a bare particle. However, because the LDOS at site n is shifted downward by $n\delta$, it follows

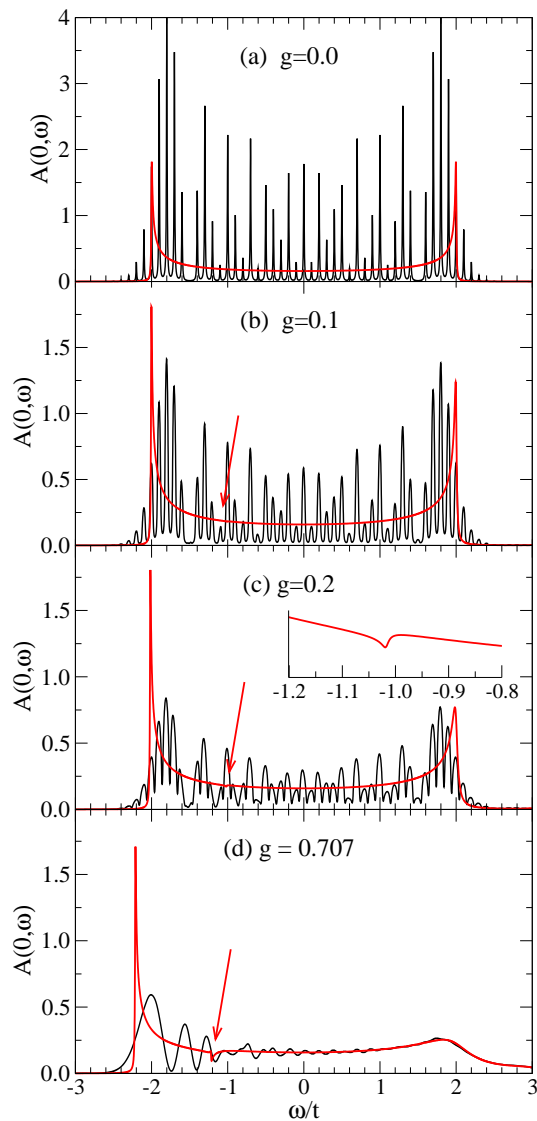


FIG. 2. (color online) $A(0, \omega)$ vs ω for $\lambda = 0, 0.005, 0.02$ and 0.25 and $t = 1, \Omega = 1, \eta = 0.005$. The thin black line shows results for $\delta = 0.1\Omega$ while the thick red line is for $\delta = 0$. Arrows mark the top of the polaron band, also see inset.

that such localized WS states could tunnel into the continuum that appears at the same energies for sufficiently large $n > 0$. In other words, such states cannot be localized, instead they are at most resonances with a width controlled by the tunneling rate. If this is large compared to δ then the resonances merge into a smoother LDOS, as we see for these parameters. Indeed, as shown below, individual resonances spaced by δ can be recovered either by increasing δ , and/or by increasing the e-ph coupling, which makes the polaron very heavy and therefore greatly decreases its tunneling rate.

Before looking at other parameters, we compare the results of SCBA with those of the other two variational approximations discussed. This comparison is shown in Fig. 3 for two values of g . For the lower value we see very good agreement between all three curves, confirming that here it is indeed very unlikely for the electron to

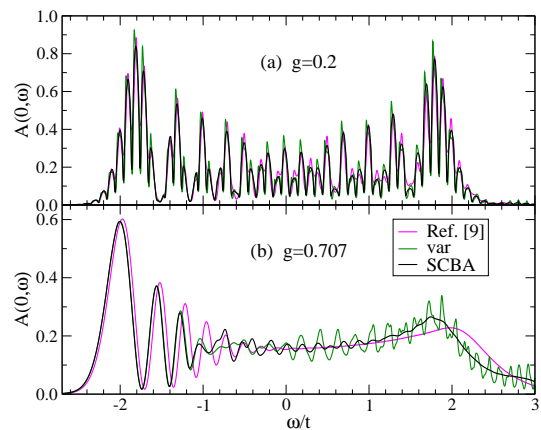


FIG. 3. (color online) Comparison between the SCBA LDOS and those predicted by the “Ref. 9” and “var” approximations (see text for more details), for parameters as in Fig. 2.

return past emitted phonons. As g increases, however, the approximation of Ref. 9 becomes less accurate, while the variational approximation which allows the electron to move to the left of the existing phonons but not to emit other phonons there, is still extremely accurate at low energies (here the two curves are indistinguishable). This shows that it is not likely for the electron to return and emit more phonons to sites where it already emitted phonons in the past, validating our assumption that sites with multiple phonons are very few for these parameters.

In Fig. 4 we show results for similarly small e-ph couplings but a much larger bias $\delta = \Omega$. Here, the broadening of the former WS states into resonances as the e-ph is turned on is very clearly visible, with their width increasing with λ . Because δ is so large, these resonances have not yet merged into a continuum even at higher energies (this occurs at larger e-ph coupling, as shown below, but larger λ is not reliably described by SCBA). The comparison with the other two variational approximations, shown in panel (d), again confirms better agreement with the assumption that the electron is free to move everywhere so long as it does not emit more phonons to the left of the last emitted one.

The results in panels (b) and (c) should be compared with the two lower curves in Fig. 4 of Ref. 9, which plot the current (not the LDOS) vs ω in a smaller range $\omega \in [-0.7, 1.7]$, and also show gaps around $\omega = \pm 0.5, 1.5$ that decrease with increasing λ . Their gaps are smaller, and in fact are nearly closed for $g = 0.5$ in agreement with the results of panel (d), which compares the three approximations. Panel (d) suggests that the variational approximation of Ref. 9 overestimates the tunneling rate resulting in broader peaks, although we must note that unlike SCBA, in Ref. 9 sites with multiple phonons are treated with the proper normalization factors. Despite these fairly minor quantitative differences, however, it is clear that qualitatively all three approximations describe similar behavior, increasing our confidence that the exact solution is not too different.

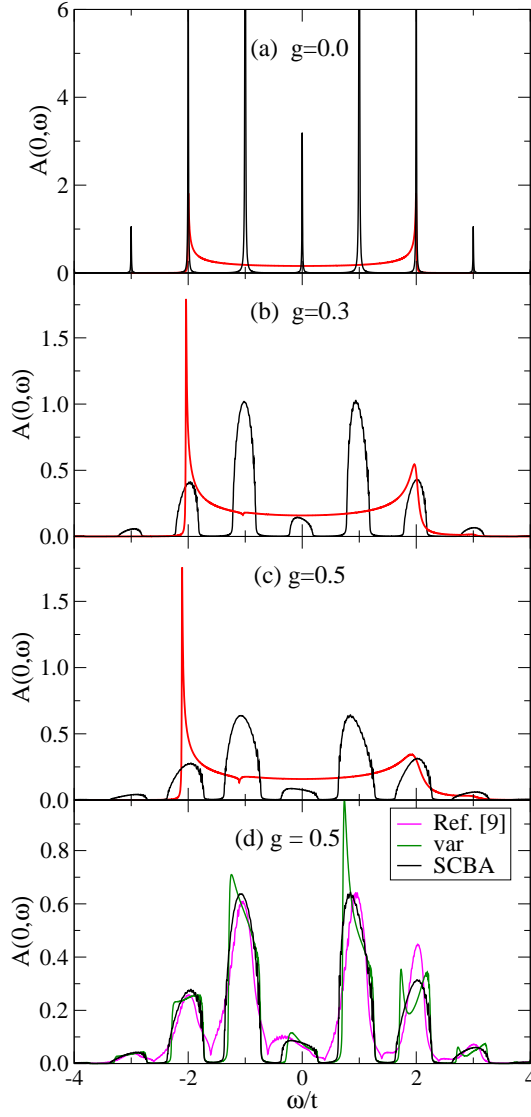


FIG. 4. (color online) Panels (a)-(c) show $A(0, \omega)$ vs ω for $\lambda = 0, 0.045$ and 0.125 and $t = 1, \Omega = 1, \eta = 0.005$. The thin black line shows results for $\delta = \Omega$ while the thick red line is for $\delta = 0$. Panel (d) compares the three approximations (see text for details).

B. Moderate and strong coupling: MA+SCBA

We now turn on the e-ph coupling and use the MA+SCBA method to study the results (for the weak couplings discussed previously, there is no difference between the MA+SCBA vs the SCBA results, as expected since at weak couplings no robust phonon cloud forms).

In Fig. 5, we show results for the small bias $\delta = 0.1\Omega$ but much larger λ values. Consider first the results in the un-biased case (thick red lines), which now show the polaron band moving towards lower energies and becoming narrower, as λ increases, as expected since the polaron becomes more stable but heavier. In panels (c) and (d), for $\lambda = 1.5$ and 2 respectively, the band associated with the second bound state²¹ is also visible below the continuum.¹²

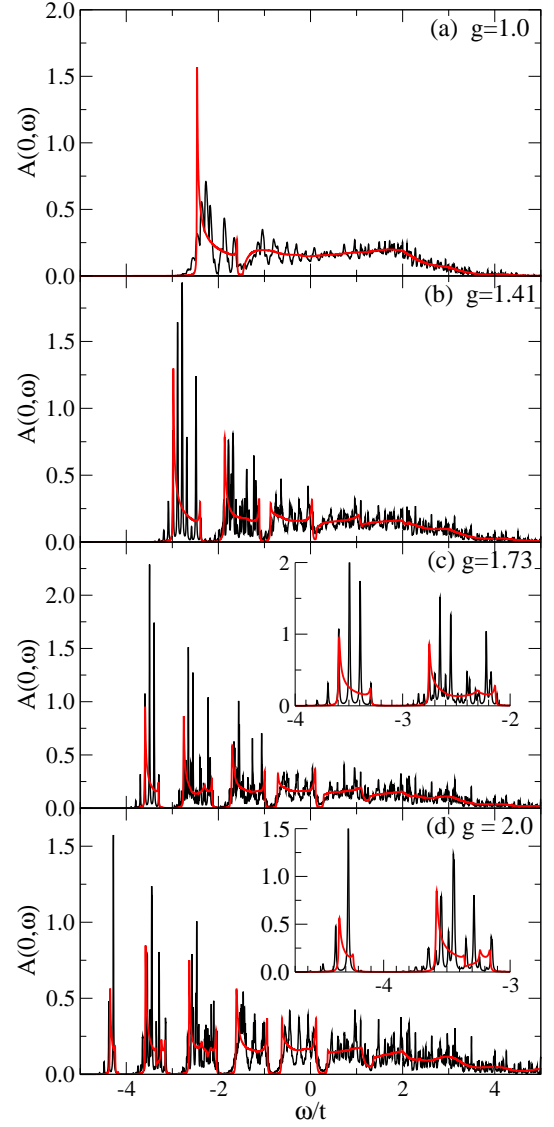


FIG. 5. (color online) LDOS for the small bias $\delta = 0.1\Omega$ when $t = \Omega = 1$, but much larger e-ph couplings $\lambda = 0.5, 1, 1.5, 2$. The thick red lines show the LDOS for $\delta = 0$. The insets zoom into the low-energy sectors.

For a finite bias, the MA+SCBA results confirm our expectations discussed above, namely that for heavier polarons the tunneling rates are significantly decreased since moving towards right to tunnel into the continuum becomes a very slow and therefore much less likely process. Indeed, for the larger λ values these tunneling rates are so small that the spectrum (at energies corresponding to the polaron band) looks like a WS ladder with the proper spacing δ between resonances, as seen more clearly in the insets. At higher energies the LDOS mimics the unbiased LDOS somewhat better, although it still has significant “peaky” structure due to tunneling out of the resonances lying further uphill.

In Fig. 6 we compare the MA+SCBA results for $\lambda = 2$ with the other approximations described above. In particular, in panel (a) we compare the low-energy sector of the $n = 0$ LDOS to that predicted by the dressed

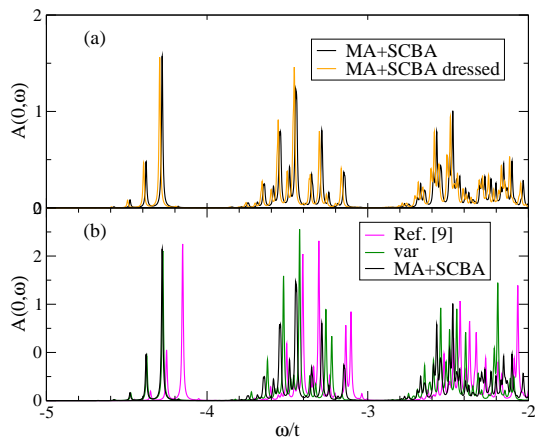


FIG. 6. (color online) Comparison between MA+SCBA and the various other approximations described in the text, for $\lambda = 2$ and $10\delta = \Omega = t = 1$.

MA+SCBA approximation. The two curves are very similar apart from a tiny shift due to the further renormalization of the polaron cloud allowed by the dressed approximation, which lowers its energy. However, it is clear that this is a very small effect, validating the assumption that a description of the phonon configuration in terms of a one-site polaron cloud plus a string of phonons left behind so that the polaron can lower its energy is reasonable. Panel (b) shows the predictions of the other two approximations, in very good agreement with MA+SCBA, at least at lower energies. This is not surprising since while the robust polaron cloud is present the electron is not expected to spend much time away from the cloud site, therefore additional restrictions on its motion should indeed have little consequences.

Finally, in Fig. 7 we show results for cases with large bias $\delta = \Omega$ and strong coupling of up to $\lambda = 2$. Individual resonances associated with different WS-like states again become visible at larger λ (in particular, see feature appearing at Ω below the polaron band) but are much broader than for the small bias. This agrees with the trends observed at weak couplings, and is expected since a larger bias must lead to increased tunneling rates even for these heavy polarons.

Comparison between the different approximations, displayed in panel (d), again shows good agreement. This suggests that the assumption implemented in SCBA to describe the phonons left behind, as the polaron moves further downhill, is still reasonable for a bias $\delta \sim \Omega$. In other words, a phonon is left behind every few sites, with low probability for multiple phonons left at the same site or for phonons emitted later to be to the left of phonons emitted earlier. For significantly larger bias δ one expects this assumption to start to fail, since in this case the carrier will need to emit many phonons at each site in order to lower its energy enough to be able to delocalize effectively. As a result, such cases cannot be described accurately by the approximations we presented here.

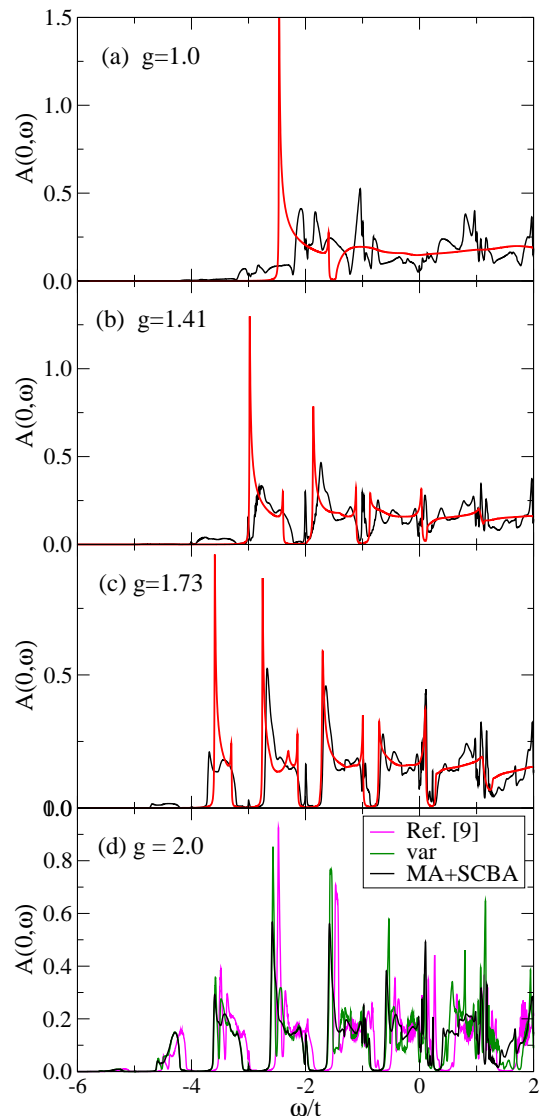


FIG. 7. (color online) LDOS for the large bias $\delta = \Omega$ when $t = \Omega = 1$, but much larger e-ph couplings $\lambda = 0.5, 1, 1.5, 2$. The thick red lines in panels (a)-(c) show the LDOS for $\delta = 0$. Panel (d) also shows the predictions of the other approximations discussed in the text.

IV. SUMMARY AND DISCUSSIONS

To summarize, we have implemented the SCBA approximation to describe the string of phonons left behind by a carrier in a biased system, in order to lower its energy to become delocalized. We argued that SCBA should provide a reasonable description for these processes if the bias is not too large. Increased coupling, however, also results in the dressing of the carrier by a phonon cloud that accompanies it as it moves through the system. Here we used the simplest variational MA flavor to describe this cloud, combining it with SCBA to describe the phonons left behind. We also showed how this formalism can be modified to implement various other variational guesses that one might want to test, and used two possible versions to validate our hypotheses for certain parameter

ranges.

Our results allow us to study the evolution of the spectrum as the bias and/or the e-ph couplings are turned on. It is worth noting that this Hamiltonian is rather unusual in that it has an unbounded spectrum if the chain is infinite: moving further along the chain will lower the energy arbitrarily much. However, we can calculate the LDOS and use it to understand the states available in the vicinity of one site. This can then be combined with the knowledge that at other sites the LDOS looks similar, apart from the appropriate energy shift, to gain a global understanding of its evolution.

We find that e-ph coupling always delocalizes the carrier, although for large coupling and small biases one can observe sharp peaks in the spectrum, that may be mistaken for localized states. As we argue, they are in fact resonances because of tunneling into delocalized states available further downhill.

While this method has been used here to study a clean 1D chain, both SCBA and MA can be straightforwardly generalized to higher dimensions, allowing this formal-

ism to be used to investigate problems that become progressively more difficult to study by numerical means.¹² Other types of e-ph coupling can also be studied by similar means,¹⁹ so that one could also investigate the relevance of the detailed modeling of the coupling to the lattice, on the behaviour of the carrier. Finally, addition of Anderson disorder is also straightforward to implement in this approach, and would open a way to investigate the competition between the localization promoted by disorder and the delocalizing effects of the e-ph coupling, away from perturbational regimes. Indeed, we believe that the method we have proposed and developed here can be used to study efficiently yet quite accurately a varied range of interesting problems.

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- ¹ for a review, see G. Nenciu, *Rev. Mod. Phys.* **63**, 91 (1991).
 - ² For eg, see E. E. Mendez, F. Agull-Rueda, and J. M. Hong, *Phys. Rev. Lett.* **60**, 2426 (1988); S. R. Wilkinson, C. F. Bharucha, K. W. Madison, Qian Niu, and M. G. Raizen, *Phys. Rev. Lett.* **76**, 4512 (1996).
 - ³ If tunneling to other bands is included, these states are resonances rather than truly localized eigenstates, see M. Glück, A. R. Kolovsky and H. J. Korsch, *Phys. Rep.* **366**, 103 (2002). Here we assume that the broadening due to Landau-Zener tunneling into other bands is negligible.
 - ⁴ G. H. Wannier, *Phys. Rev.* **117**, 432 (1960).
 - ⁵ R. Landauer, *Philos. Mag.* **21**, 863 (1970).
 - ⁶ D. Emin and C. F. Hart, *Phys. Rev. B* **36**, 2530 (1987).
 - ⁷ For a review, see C. Deibel and V. Dyakonov, *Rep. Prog. Phys.* **73**, 096401 (2010) and references therein.
 - ⁸ Work on clean organic semiconductors is reported, for example, in I. G. Lezama *et al*, *Nat. Mat.* **11**, 788 (2012).
 - ⁹ J. Bonca and S. Trugman, *Phys. Rev. B* **79**, 4874 (1997).
 - ¹⁰ W. Zhang, A. O. Govorov and S. E. Ulloa, *EuroPhys. Lett.* **58** 857 (2002); *ibid*, *Phys. Rev. B* **66**, 134302 (2002).
 - ¹¹ L. Vidmar *et al*, *Phys. Rev. B* **83**, 134301 (2011).
 - ¹² M. Berciu, *Phys. Rev. Lett.* **97**, 036402 (2006); G. L. Goodvin, M. Berciu, and G. A. Sawatzky, *Phys. Rev. B* **74**, 245104 (2006).
 - ¹³ T. Holstein, *Ann. Phys. (NY)* **8**, 325 (1959); *ibid*, **8**, 343 (1959).
 - ¹⁴ G. D. Mahan, *Many particle physics*, 3rd ed. (Kluwer Academic, New York, 2000).
 - ¹⁵ E. N. Economou, *Green's functions in Quantum Physics*, 3rd ed. (Springer-Verlag, New York, 2006).
 - ¹⁶ H. Fukuyama, R. A. Bari and H. C. Fogedby, *Phys. Rev. B* **8**, 5586 (1973).
 - ¹⁷ S. G. Davison, R. A. English, Z. L. Miskovic, F. O. Goodman, A. T. Amos and B. L. Burrows, *J. Phys.: Condens. Matter* **9**, 6371 (1997).
 - ¹⁸ O. S. Barisic, *Phys. Rev. Lett.* **98**, 209701 (2007); M. Berciu, *Phys. Rev. Lett.* **98**, 209702 (2007); M. Berciu and G. L. Goodvin, *Phys. Rev. B* **76**, 165109 (2007).
 - ¹⁹ G. L. Goodvin and M. Berciu, *Phys. Rev. B* **78**, 235120 (2008); D. Marchand, G. De Filippis, V. Cataudella, M. Berciu, N. Nagaosa, N. V. Prokof'ev, A. S. Mishchenko, and P. C. E. Stamp, *Phys. Rev. Lett.* **105**, 266605 (2010); F. Herrera, K. W. Madison, R. V. Krems, M. Berciu, *Phys. Rev. Lett.* **110**, 223002 (2013).
 - ²⁰ Note that the number of phonons at site n_k is not necessarily $p+1$, since in principle it is possible that the electron has been at site n_k before and created a phonon, after which it moved to create phonons at other sites and only then returned to n_k to create a cloud. As already argued such processes are not very likely. To the extent that they happen, their effect is to replace $p \rightarrow p+1$ if one extra phonon was already there, etc. For large couplings where the typical number p of phonons in the cloud can be large, this will have relatively small numerical consequences.
 - ²¹ J. Bonca, S. A. Trugman and I. Batistic, *Phys. Rev. B* **60**, 1633 (1999).